Early unusual ozone loss during the Arctic winter 2002/2003 compared to other winters

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F. Goutail et al.

Abstract

Total column ozone reduction in the Arctic is evaluated each winter since 1993/1994 by the transport method (3-D CTM passive ozone minus measurements). The cumulative loss from 1 December to the end of the season ranges from 5–10% during warm winters like 1998/1999, 2000/2001 and 2001/2002 up to 30%–32% during cold winters like 1994/1995 and 1995/1996. The 23% cumulative loss observed during the winter 2002/2003 is similar in amplitude to the 20–24% measured in 1996/1997 and 1999/2000 but the timing is different. It started unusually early in December after the occurrence of very low temperature at all stratospheric levels between 550 K and 435 K allowing PSC formation and thus chlorine activation. The early ozone loss of 2002/2003 is well captured by current 3-D CTM models.

1. Introduction

Chemical ozone losses in the Arctic regions have been studied since the early 1990s. Because of the large activity of planetary waves in the northern hemisphere, the evaluation of the chemical loss is not straightforward. It requires precise removal of the transport component. Several methods have been suggested, described and intercompared by Harris et al. (2002). One of these is the transport model which consists in comparing measurements with a 3-D model in which ozone is considered as a passive tracer (Goutail et al., 1999).

After presenting the measurements and the models used in this study, the ozone loss for the winter 2002/2003 will be evaluated and analyzed. The results will be compared to the loss calculated by the same method for all previous winters since 1993/1994. Finally, the ability of two 3-D chemical transport models (CTMs) to reproduce the early unusual ozone loss observed in 2002/2003 will be investigated.

2. Measurements and models

The data used here are total ozone columns measured at the seven Arctic stations equipped with SAOZ UV-visible spectrometers (Pommereau and Goutail, 1988) part of the Network for the Detection of Stratospheric Changes (NDSC), plus those of a similar spectrometer but of different design operated at Harestua in Southern Norway (Van Roozendael et al., 1995). The location of the stations, the date of installation and the institutes running the instruments are displayed in Table 1. Among those spectrometers, one is a little north of the polar circle (Scoresbysund) and therefore starts observing only in mid-January, and two are at higher latitude (Thule and NyAlesund) beginning measuring only in mid-February.

Ozone slant columns are measured every morning and evening between 86° and 91° solar zenith angle (SZA) in the visible Chappuis bands where the absorption cross sections are well known (1% uncertainty) and insensitive to temperature (<1%). Total ozone is retrieved from slant column densities using a constant Air Mass Factor (AMF) calculated for a typical Arctic ozone profile measured by the balloon-borne version of the SAOZ instrument. The AMF at 90° SZA is 16.5. AMF fluctuations related to changes in shape of ozone profile are smaller than 3% (Sarkissian et al., 1995). The consistency of the measurements of the various instruments demonstrated during inter-comparison campaigns is of ±3% (Vaughan et al., 1997; Roscoe et al., 1999). The presence of dense high type II polar stratospheric clouds (PSCs), which may lead to an underestimation of total ozone because of the lifting of the scattering layer, is detected by looking at a colour index (Sarkissian et al., 1991) and the corresponding ozone data are removed.

The CTM models used here are REPROBUS and SLIMCAT run in passive and photochemical modes. Though using similar approaches, they differ in several aspects which need to be understood before discussing possible differences in the results. The two models are described in Appendix A and B. To calculate the ozone loss from SAOZ, the transported passive ozone from REPROBUS is used. For the winter 2002/2003, both REPROBUS and SLIMCAT were initialized on 1 December 2002.

The study is limited to the polar stratospheric vortex. The selection of data is based on potential vorticity (PV) fields calculated by the Danish Meteorological Institute from ECMWF analyses. The boundary of the vortex is given by the maximum PV gradient following the procedure suggested by Nash et al. (1996). This procedure defines three limits of the vortex, the "equatorward boundary", the "vortex edge", and the "poleward boundary". Here, the "poleward boundary" on the 475 K isentropic surface (approx. 17 km – maximum of ozone concentration) is chosen. The broad geographical distribution of the stations means that one or more are typically located in the vortex on a given day during the winter period.

3. Ozone loss above individual SAOZ stations in 2002/2003

Figure 1 shows the evolution of the ozone column and the position of the vortex at one of the Arctic stations, Sodankyla in Northern Finland, between December 2002 and April 2003. The upper panel displays observed (shaded area) and passive ozone simulated by REPROBUS (solid black line). The lower panel shows PV at 475 K above the station and the average PV at the edge of the vortex. Modeled and measured column ozone are comparable. The model captures well the short-term fluctuations related to the reversible vertical motions of the tropopause following the propagation of planetary waves. Transitions between the inside and the outside of the vortex are also clearly seen, specially during the second half of January, when the station is located outside the vortex and high ozone columns up to 500 DU are measured. However, a significant difference progressively builds up indicative of a chemical reduction. Just before the end of the simulation, by mid-March, when the vortex passes for the last time above Sodankyla, the difference between the measurements and the REPROBUS passive ozone reaches 90 Dobson Units (DU), or about 23%.

Similar observations are reported at the other stations (not shown) though the total ozone and hence the absolute loss expressed in DU can vary from one station to the

ACPD

4, 5019–5044, 2004

other because of the change of tropopause height. The loss at the various stations should thus be compared in relative and not absolute units.

The relative percent reduction in the vortex at the seven stations combined altogether is shown in the lower panel of Fig. 2. Also shown in the upper panel of Fig. 2 is the minimum ECMWF temperature north of 30° N at three levels (435, 475 and 550 K). At 550 K the temperatures were well below T_{NAT} from mid-November. At the beginning of December they were below T_{NAT} at the upper levels (475 and 550 K) and by mid-December at the three levels where they remained cold until mid-January. After that date, three minor stratospheric warming occurred, the first around 15 January lasting 20 days, the second around 10 February lasting 15 days and the last around 5 March for 10 days. During the short warming periods, the temperature increases above the T_{NAT}. The final warming is observed around 20 March.

Consistent with the meteorology, the loss started very early (Fig. 2, bottom), during the first ten days of December, at an average rate of 0.35% per day. At the end of December, a 10% total column ozone reduction is already observed above the SAOZ stations, four of them being located at (or southward) the polar circle. During the first ten days of January 2003, the temperature was still below that of T_{NAT} PSC formation but the ozone reduction remained stable. A second period of reduction occurred between 10 January and 31 January 2003 at a rate of 0.4% per day. Then it stabilized again during twenty days. A third period of reduction at a rate of 0.5% per day is observed between 20 February and 1 March 2003. After that, no additional loss is seen in the SAOZ measurements leading to a cumulative loss of 23% at the end of the winter or ~90 DU. Also shown (Fig. 2, bottom) is a 10-day mean with error bars representing the standard deviation. On average, the standard deviation is 4%.

In summary, a total cumulative ozone loss of 23±4% was observed by the SAOZ network at the end of the winter, half of which occurred in December.

ACPD

4, 5019–5044, 2004

Early unusual ozone loss during the Arctic winter 2002/2003 compared to other winters

F. Goutail et al.

4. Early chlorine activation

A very early ozone loss in 2002/2003 has been also reported by Tilmes et al. (2003) from an analysis of the HALOE data using the TRAC (ozone-tracer correlation) method, associated with very low HCl mixing ratio measured on the 520 K surface by the MkIV balloon instrument flown over northern Sweden on 16 December 2002. These low HCl mixing ratios indicate that strong chlorine activation had already occurred by that date in the vortex.

This is in agreement with PSC observed by balloon-borne instruments, during the first days of December 2002, between 3 and 7 December (Larsen et al., 2004).

The occurrence of early chlorine activation is also supported by the measurements of the ODIN Sub-Millimeter Radiometer (SMR) satellite instrument. Significant amount of ClO, around 0.3–0.6 ppbv, were measured on 9–10 December 2002, between 450 K and 525 K (Urban et al., 2004).

The early activation is also confirmed by the presence of high OClO levels observed in December by the UV-Visible spectrometer at Harestua in Southern Norway (Fig. 3) when the vortex was present above the station (bottom panel). It is also confirmed by the ERS-2/GOME (Global Ozone Monitoring Experiment on board ERS-2 satellite) observations displaying an unusual high activation in December compared to the previous winters (see: http://www.iup.physik.uni-bremen.de/gomenrt2003/)(Wagner et al. 2001, 2002; WMO assessment 200, Chapter 3).

The early ozone loss observed by the SAOZ in December 2002 is thus consistent with the cold ECMWF stratospheric temperatures, the HCl reduction observed by the MkIV balloon instrument, the high ClO of SMR/ODIN and the high OClO over Harestua as well as that reported by ERS-2/GOME.

ACPD

4, 5019–5044, 2004

Early unusual ozone loss during the Arctic winter 2002/2003 compared to other winters

F. Goutail et al.

Title Page
Abstract | Introduction
Conclusions | References
Tables | Figures
◀| |▶
◀ ▶
Back | Close
Full Screen / Esc

Print Version

Interactive Discussion

5. Comparison to other winters

The results of the winter 2002/2003 are compared in Fig. 4 to that of all other winters since 1993/1994: derived minimum temperature at 475 K on the left and SAOZ cumulative O_3 loss and of the two models on the right, modeled ozone loss being derived by difference between two consecutive runs (passive and photochemical modes). The 23% cumulative loss during the winter 2002/2003 is larger than the 5–10% observed during warm winters like 1998/1999, 2000/2001 and 2001/2002 (Goutail et al., 2000, 2003), but smaller than the 30%–32% reported during the coldest winters in 1994/1995 and 1995/1996 (Goutail et al., 1999, 1998). It is similar in amplitude to the 20–24% measured in 1996/1997 and 1999/2000 (Goutail et al., 1998; Harris et al., 2002), but the timing is different. While during most of the winters, the loss begins in January or eventually in mid-December like in 1995/1996, in 2002/2003, it started very early in December in coincidence with very low stratospheric temperatures.

Noteworthy also in Fig. 4 is that the observed loss in January 1994/1995 and January 1995/1996 is not reproduced by the REPROBUS and SLIMCAT simulations available for those winters. Goutail et al. (1999) compared SAOZ and 3-D CTM ozone loss for the winter 1994/1995. They found that the timing and the altitude at which the ozone reduction took place was well captured by the 3-D CTMs, for that winter, but that the amplitude was underestimated in low sun conditions during the early winter. Since this statement, improvements have evidently been made in the two 3-D CTMs.

The observed cumulative loss for the eleven consecutive winters since 1993/1994, the main period of ozone reduction during the season and the date at which 10% loss was reached are displayed in Table 2. In most cases, the ozone reduction is starting during the second half of December and 10% loss is reached only on or after 20 January. The winter 2002/2003 with its 10% loss at the end of December is an extreme case.

ACPD

4, 5019–5044, 2004

6. Model simulations of 2002/2003

The ozone loss above the SAOZ stations during the winter 2002/2003 has been simulated by REPROBUS and SLIMCAT. For better consistency with observations, only simulations corresponding to existing measurements (SZA <91° at noon) are considered.

6.1. REPROBUS

The REPROBUS simulations are shown in Fig. 5 in a similar format as in Fig. 2. Also shown is a 10-day average with error bars representing standard deviation. On average, the dispersion is 2%, half that of the measurements). REPROBUS captures a loss at a rate of 0.3% per day in December, then a plateau of about 20 days and again a loss at a rate of 0.2% per day from 20 January until 20 March. Its cumulative loss at the end of the winter is 20% (or 80 DU).

The timing and the amplitude of the loss are comparable to the observations. The current REPROBUS version correctly captures the December ozone destruction.

The details of simulated fields (PSC surface area, HCl, ClO_x, O_3 loss and PV) at 550 K and 475 K on 5 and 25 December and 15 January, are displayed in Fig. 6. PSCs are already present on 5 December (Fig. 6, top), more developed at 550 K, resulting in an HCl depletion in the vortex, as well as in the appearance of reactive chlorine (ClO_x=[ClO]+2[ClOOCl]). However, there is no indication of ozone loss as the vortex is confined in darkness north of 60° N.

On 25 December (Fig. 6, middle), few PSCs could be still seen, but HCl was completely depleted and chlorine largely activated in the whole vortex. However, the vortex is now elongated towards illuminated latitudes, and significant ozone losses appear at its periphery, more abundant at 475 K than at 550 K.

On 15 January (Fig. 6, bottom), no more PSCs could be seen, but the vortex is still activated and its displacement towards East European mid-latitudes as south as 50° N, allows the ozone depletion to reinforce at its inner edge, resulting in an annular shape.

ACPD
4, 5019–5044, 2004

At the end of January, (not shown), the depletion is well mixed and the vortex is homogeneously depleted, in agreement with the conclusions of Tilmes et al. (2004).

6.2. SLIMCAT

The results of SLIMCAT simulations are shown in Fig. 7. The early depletion is also captured at a rate of 0.3% per day in December and January followed by a plateau of about 10 days in February and then by a loss again at a rate of 0.3% per day from 10 February until 10 March. At the end of the winter, the cumulative loss is 20% (or 80 DU). The timing and the amplitude of the depletion are similar to that of REPROBUS. The SLIMCAT results for 2002/2003 are described in more detail in Feng et al. (2004).

In summary, the 2002/2003 REPROBUS and SLIMCAT simulations are in close agreement with each other, as well as with the SAOZ observations within the error bars. In their current versions, both models appear to capture the reported low-sun early winter ozone depletion.

7. Conclusions

An unusually early ozone loss was observed in 2002/2003, at least one month earlier than during any of the previous eleven winters. This unusual behaviour is consistent with the low reported temperatures in the stratosphere as well to the signature of early chlorine activation seen by ground-based, balloon and satellite observations. The early ozone depletion is well captured by the current versions of both the REPROBUS and SLIMCAT models, showing that chemical depletion did indeed take place in December at low sun, but predominantly at the illuminated edge of the distorted vortex.

ACPD
4, 5019–5044, 2004

Appendix A: REPROBUS

The 3-D-CTM REPROBUS (Reactive Processes Ruling the Ozone Budget in the Stratosphere) is described in Lefèvre et al. (1994, 1998). In general the model is used for seasonal simulations run at 2° latitude by 2° longitude resolution (approximately 220 km×90 km at the polar circle). The transport and chemical reactions rated are driven by the ECMWF 6-hourly analysis of temperature and winds, analyzed vertical velocities to compute vertical motion. The REPROBUS chemical package is described in Lefèvre et al. (1994) and has been regularly updated since from newly measured reaction rates or absorption cross-sections (DeMore et al., 1997; Brown et al., 1999a, b; Sander et al., 2000; Knight et al., 2002; Roehl et al., 2002). PSCs are assumed to be in thermodynamic equilibrium. The composition of liquid aerosols is calculated analytically. NAT is formed at the equilibrium NAT saturation temperature and the presence of ice is tested using the saturation vapour pressure of water over ice. Ice is assumed to incorporate NAT as a co-condensate, removing nitric acid from the vapour phase. The description of polar stratospheric cloud microphysics in the model has also been improved in recent years. In the most recent version of REPROBUS, a highly selective nucleation process allows the formation of a small number of large NAT (Nitric Acid Trihydrate) particles, as observed from airborne measurements in winter 1999–2000 (Fahey et al., 2001). These particles may form at temperatures above the ice frost point, and may coexist with liquid ternary aerosols. Above the NAT formation threshold temperature, these solid particles are able to re-evaporate.

The ozone fields are initialized every year at the beginning of December from MLS or POAM measurements, depending upon their availability. The three-dimensional ozone analysis made at ECMWF was used to initialize the 2002/2003 simulation. Water vapour is initialized from a MLS-HALOE zonal mean late fall climatology. The sulfate aerosol content is derived for each winter from SAGE-II measurements. All other species are initialized from the November zonal mean of a 5-year run of REPROBUS coupled to the ARPEGE general circulation model (see WMO 1998 assessment, Chapter 12).

REPROBUS simulations used in the present work are those carried out every year with the latest version available at that time. Until the end of 1998, the REPROBUS calculations were performed on the 31 vertical levels of the ECMWF analysis, extending up to 10 hPa at that time. In January 1999, following the extension of the vertical domain of the ECMWF analyses to 60 levels up to 0.1 hPa, the number of vertical levels of REPROBUS was increased to 42, with a ~1 km vertical resolution in the lower stratosphere and an uppermost level at 0.1 hPa (about 65 km). The earliest integrations of the model, prior to 1999, for which the 60-level ECMWF operational analysis are not available, could not be reprocessed with the last version of the model. The REPROBUS initialization date, model run number and number of vertical levels are summarized in Table 3.

Appendix B: SLIMCAT

The 3-D SLIMCAT CTM, is described in detail by Chipperfield (1999) and has been used for a number of studies related to ozone depletion in the northern and southern vortices (e.g. Chipperfield et al., 1996, 1998; Guirlet et al., 2000; Hansen et al., 1997; Sinnhuber et al., 2000). In general the model is run for multiannual simulations at low horizontal resolution ($7.5^{\circ}\times7.5^{\circ}$) and these are used to initialize higher resolution seasonal simulations.

For past studies seasonal integrations of SLIMCAT were run at a resolution of 2.5° latitude $\times3.75^{\circ}$ longitude (approximately 275 km×170 km at the polar circle) using UK Met Office (UKMO) horizontal winds and temperatures. These runs used 24θ (isentropic) levels from 330 K to 3000 K (approximately 10 to 55 km) and the vertical advection was derived from calculated heating rates using the MIDRAD stratospheric radiation scheme (Shine, 1987). The seasonal runs were initialized from the output of a UKMO-forced multiannual integration which started in October 1991. The initialization occurred on 1 November for all winters before 1999 and on 1 December after,

ACPD
4, 5019–5044, 2004

Early unusual ozone loss during the Arctic winter 2002/2003 compared to other winters

F. Goutail et al.

in order to match the initialization date of REPROBUS. For these runs a tropospheric contribution was added to the SLIMCAT O_3 column in order to compare with SAOZ observations. This can cause significant uncertainties in periods of large variation of the tropopause height.

Recently SLIMCAT has been modified to use a hybrid $\sigma-\theta$ coordinate and extended down to the surface. Here this most recent version of the model has been used to provide the seasonal simulations for the winters 1999/2000, 2002/2003 and 2003/2004 (the other winters use results from the older model described above). These runs were forced by ECMWF analyses and had a horizontal resolution of 2.8°×2.8° with 24 levels from the surface to ~55 km. They were initialized from an ECMWF-forced multiannual run which started in 1989 (run 317). These seasonal model runs are described and used in Feng et al. (2004).

The SLIMCAT gas-phase and heterogeneous chemistry modules, including liquid aerosols, NAT and ice particles, are described in Chipperfield (1999). PSCs are assumed to be in thermodynamic equilibrium, with no modification of the model temperature to account for supersaturation. The composition of liquid aerosols (containing HNO_3, H_2SO_4, H_2O and HCl) is calculated analytically. NAT is formed at the equilibrium NAT saturation temperature and the presence of ice is tested using the saturation vapour pressure of water over ice. Ice is assumed to incorporate NAT as a co-condensate, removing nitric acid from the vapor phase. The older (UKMO-forced) runs used an ice-based denitrification scheme, which had essentially no effect in the Arctic simulations used here. The recent (ECMWF-forced) runs incorporated a simple NAT-based denitrification scheme (see Davies et al. 2001; Feng et al., 2004).

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ACPD

4, 5019–5044, 2004

Early unusual ozone loss during the Arctic winter 2002/2003 compared to other winters

F. Goutail et al.

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Table 1. Arctic ground-based SAOZ NDSC stations.

Location	Lat.	Long.	Since	Institute
Ny-Alesund, Svalbard	79 N	12 E	1991	NILU
Thule, West Greenland	77 N	69 W	1991	DMI
Scoresbysund, Greenland	70 N	22 W	1991	CNRS/DMI
Zhigansk, East Siberia	67 N	123 E	1991	CNRS/CAO
Salekhard, West Siberia	67 N	67 E	1998	CNRS/CAO
Sodankyla, Finland	67 N	27 E	1990	CNRS/FMI
Harestua, Norway	60 N	11 E	1994	IASB

ACPD

4, 5019–5044, 2004

Table 2. SAOZ Ozone loss.

Winter	Loss (%)	Loss (DU)	Period of loss	10% loss reached
1993/1994	17	75	20 Dec.–20 Mar.	20 Jan.
1994/1995	31	140	20 Dec.–20 Mar.	20 Jan.
1995/1996	30	125	20 Dec.–10 Mar.	20 Jan.
1996/1997	22	95	30 Jan.–30 Mar.	20 Feb.
1997/1998	17	80	10 Dec.–1 Mar.	10 Feb.
1998/1999	5	25	1 Jan.–20 Feb.	–
1999/2000	23	105	1 Jan.–10 Mar.	10 Feb.
2000/2001	10	52	10 Jan.–1 Mar.	10 Mar.
2001/2002	10	46	10 Dec.–30 Mar.	30 Mar.
2002/2003	23	90	1 Dec.–10 Mar.	30 Dec.
2003/2004	18	90	1 Jan.–1 Mar.	30 Jan.

ACPD
4, 5019–5044, 2004

Table 3. REPROBUS model run.

Winter	Initialization date	Model run	Vertical levels	Upper level
1993/1994	1 Dec. 1993	200	31	10 hPa
1994/1995	28 Nov. 1994	004	31	10 hPa
1995/1996	1 Dec. 1995	152	31	10 hPa
1996/1997	26 Dec. 1996	310	31	10 hPa
1997/1998	1 Dec. 1997	505	31	10 hPa
1998/1999	23 Dec. 1998	604	31	10 hPa
1999/2000	1 Dec. 1999	814	42	0.1 hPa
2000/2001	1 Dec. 2000	900	42	0.1 hPa
2001/2002	1 Dec. 2001	911	42	0.1 hPa
2002/2003	1 Dec. 2002	1101	42	0.1 hPa

ACPD
4, 5019–5044, 2004

Early unusual ozone loss during the Arctic winter 2002/2003 compared to other winters

F. Goutail et al.

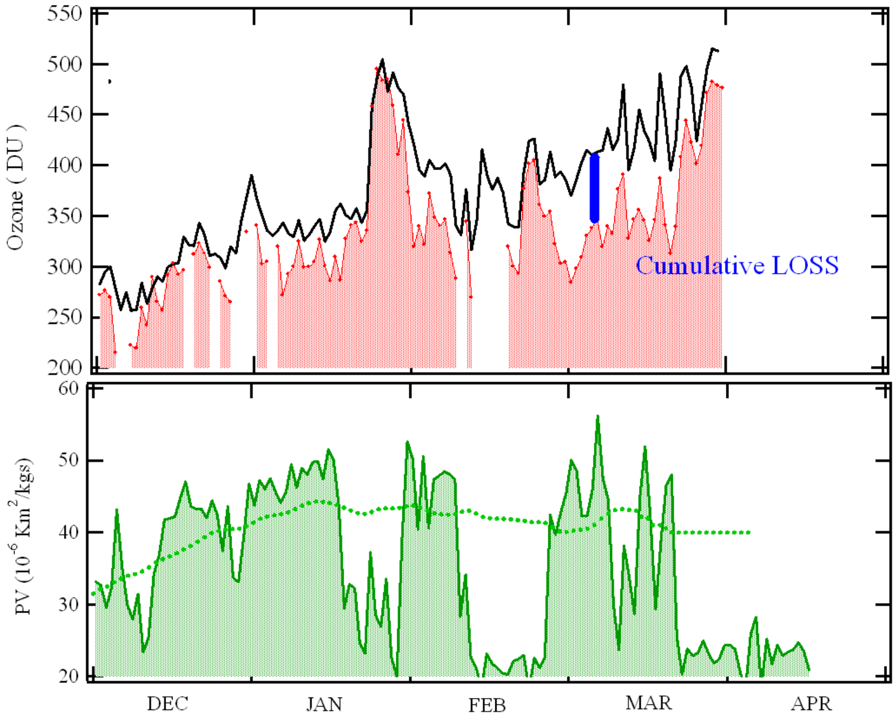

Fig. 1. Top: time series of total ozone measured (pink shaded area) and simulated by the transport model REPROBUS (thick line) from 1 December 2002 until 30 April 2003 above Sodankyla, Finland. Bottom: potential vorticity at 475 K (green shaded area) and limit of the vortex (green thick line).

ACPD
4, 5019–5044, 2004

Early unusual ozone loss during the Arctic winter 2002/2003 compared to other winters

F. Goutail et al.

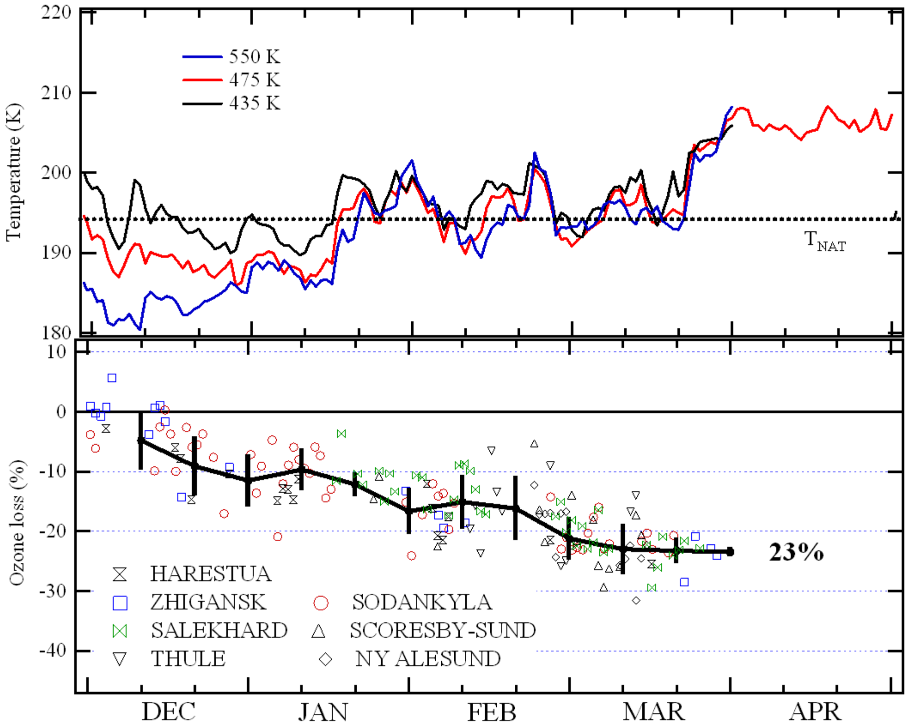

Fig. 2. Top: ECMWF minimum temperature in the Arctic at 3 levels, 550 K, 475 K and 435 K. Bottom: measured total ozone reduction inside the vortex at the SAOZ Arctic stations (symbols), 10-day average (solid line) and standard deviation (error bars) from 1 December 2002 until 30 March 2003.

ACPD

4, 5019–5044, 2004

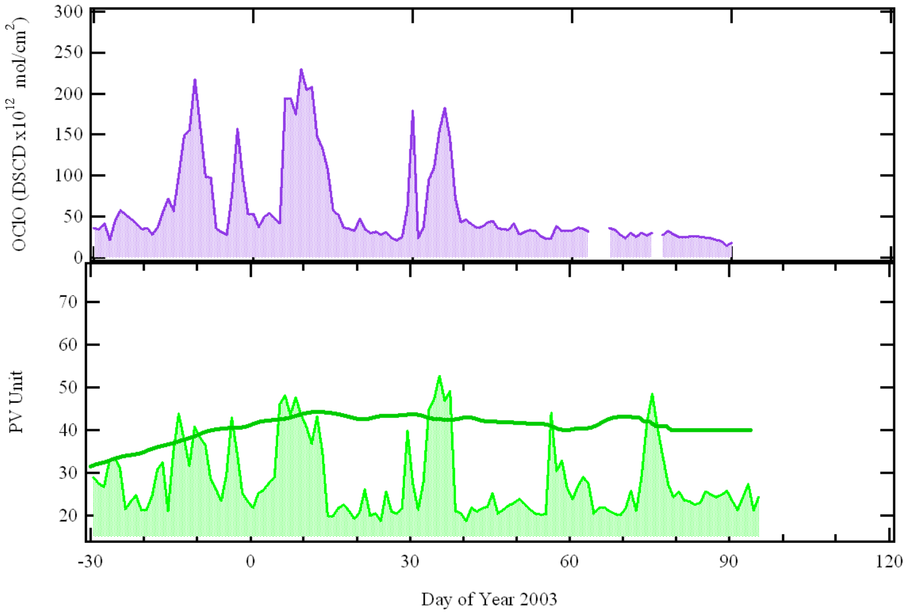

Fig. 3. Top: time series of OClO Differential Slant Columns (91°–80° SZA) at Harestua, southern Norway, from 1 December 2002 until 30 April 2003. Bottom: potential vorticity at 475 K and limit of the vortex.

ACPD
4, 5019–5044, 2004

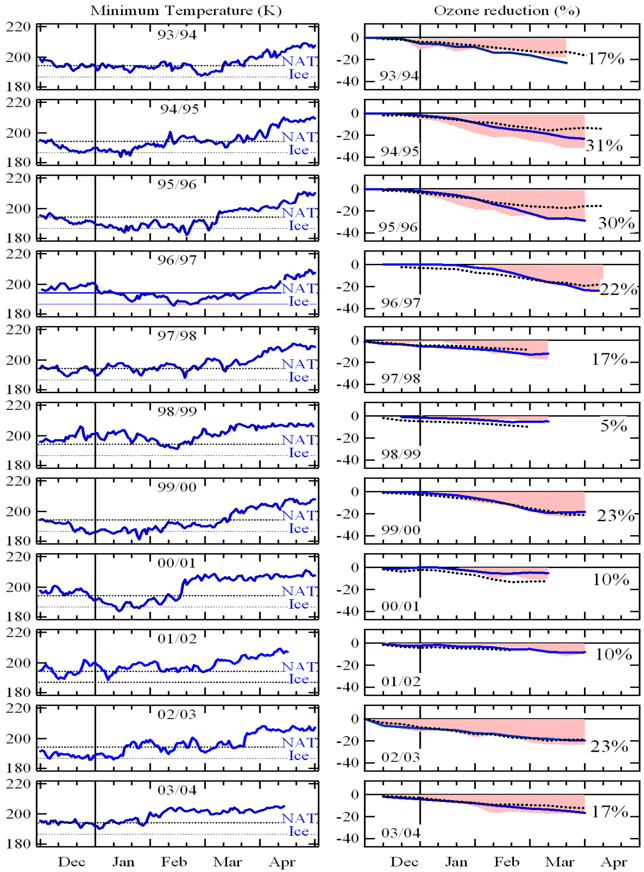

Fig. 4. Right: total ozone reduction in the vortex derived each year since 1993/1994 from the measurements of the SAOZ stations (shaded pink) and simulated by REPROBUS (blue solid line) and SLIMCAT (blue dotted line). Left: minimum temperature at 475 K north of 30° N and limit for NAT and ice PSC formation.

ACPD
4, 5019–5044, 2004

Title Page
Abstract
Introduction
Conclusions
References
Tables
Figures
Back
Close
Full Screen / Esc
Print Version
Interactive Discussion

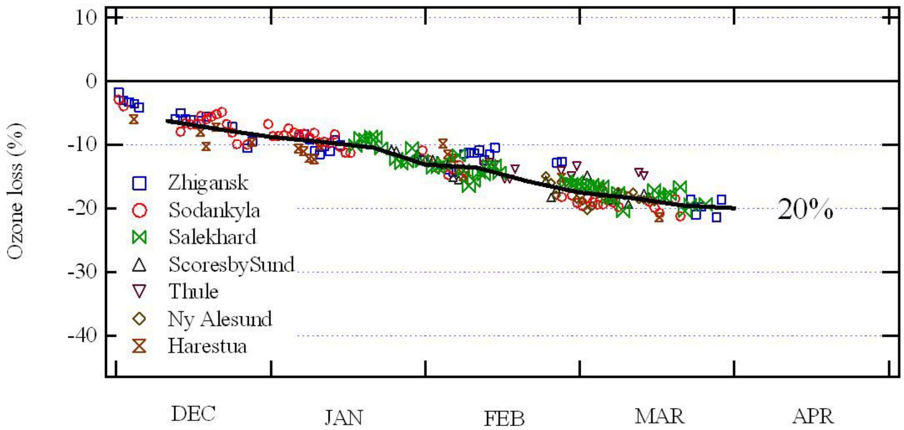

Fig. 5. REPROBUS ozone loss inside the vortex at the SAOZ Arctic stations (symbols), 10-day average (solid line) and standard deviation (error bars) from 1 December 2002 until 30 March 2003.

ACPD

4, 5019–5044, 2004

Early unusual ozone loss during the Arctic winter 2002/2003 compared to other winters

F. Goutail et al.

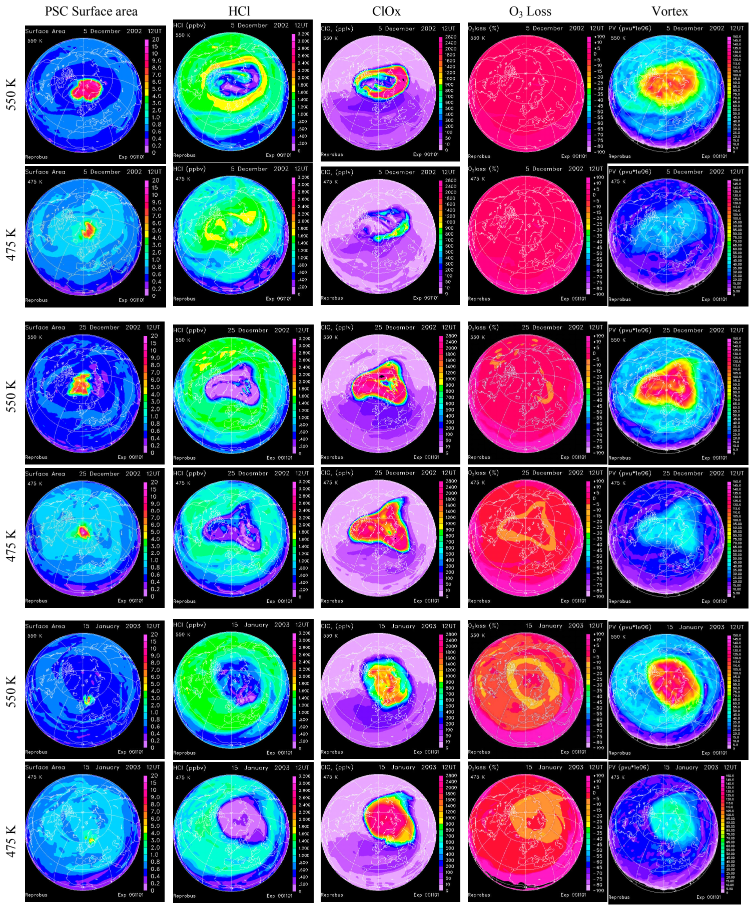

Fig. 6. PSC, HCl, ClO_x, O_3 loss and vortex surface area from REPROBUS at two isentropic levels 550 K and 475 K (colour scale – ClO_x: pink 2000 pptv – O_3 loss: orange 5–10%, yellow 10–15%); top: on 5 December 2002, middle: on 25 December 2002, bottom: on 15 January 2003.

ACPD

4, 5019–5044, 2004

Early unusual ozone loss during the Arctic winter 2002/2003 compared to other winters

F. Goutail et al.

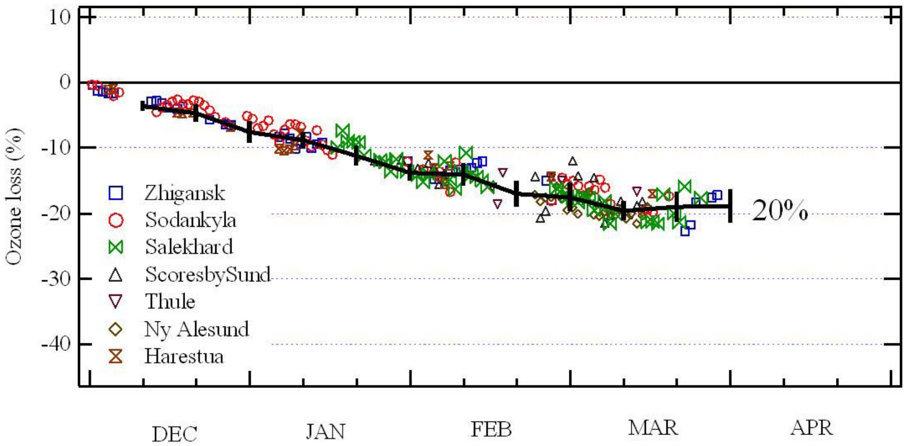

Fig. 7. Same as Fig. 5 but for SLIMCAT.

ACPD
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Early unusual ozone loss during the Arctic winter 2002/2003 compared to other winters

F. Goutail et al.